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Characterization of a low-energy constricted-plasma source

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Abstract

The construction and principle of operation of the Constricted-Plasma Source are described. A supersonic plasma stream is produced by a special form of a dc-glow discharge, the constricted glow discharge. The discharge current and gas flow pass through an orifice of small diameter (constriction) which causes a space charge double layer but also serves as a nozzle to gasdynamically accelerate the plasma flow. Plasma parameters have been measured using Langmuir probes, optical emission spectroscopy, and a plasma monitor for mass-

resolved energy measurements. Experiments have been done with nitrogen as the discharge gas. It was found that the energy distribution of both atomic and molecular ions have two peaks at about 5 eV and 15 eV, and the energy of almost all ions is less than 20 eV. The ionization efficiency decreases with increasing gas flow. The downstream plasma density is relatively low but activated species such as excited molecules and radicals contribute to film growth when the source is used for reactive film deposition.

I. INTRODUCTION

Surface processing of materials is often assisted by, or even requires the use of a suitable plasma or ion beam source. Sources of streaming plasma (also referred to as "downstream plasma sources") can be viewed as high-current, fully space-charge-compensated, very low-energy ion sources. All of these features are desired for the deposition of thin films, in particular crystalline films. Ion sources as well as downstream plasma sources are categorized by their discharge type, for example dc filament (Kaufman) sources ¹, RF sources ², microwave sources ³, electron cyclotron resonance (ECR) sources ⁴, Hall effect ion sources ⁵, Penning sources ^{6, 7}, thermionic arc sources (plasmatrons) ⁸, etc. For some processes, not necessarily ion beams or streaming plasma are used but flows of activated species such as radicals or atoms of molecular gases ⁹⁻¹². Activated species are important in the deposition of many thin films systems; they are produced along with the charged plasma particles (ions, electrons) and are therefore listed here as well.

In the present paper we characterize a relatively unknown kind of downstream plasma source, the constricted-plasma source. Early versions of this source have been introduced by Miljevic and applied as either spectroscopic light sources ¹³ or, in combination with particle extraction optics, as sources of energetic ions ¹⁴ and electrons ¹⁵. We'll describe the construction, principle of operation, performance, advantages and disadvantages, and some applications of the source.

II. CONSTRUCTION AND PRINCIPLE OF OPERATION

The section is an extension of an earlier paper, the "Working Principle of the Hollow-Anode Plasma Source" ¹⁶. To avoid confusion with names ("constricted" versus "hollow-anode"), we need to look at the principal source construction (Fig. 1) and its modes of

operation.

Let's consider a dc-glow discharge which is typically current-limited by either an external resistor or by the current mode of a modern electronically switched supply. In contrast to a conventional glow discharge, the active anode area is intentionally reduced by a blocking insulator to only some mm² as shown Fig. 2(a). The current density at the available anode area is unusually high, of order 0.1 A/mm². This current density cannot be carried by the pre-anode plasma unless an electric double layer forms which acts effectively as a virtual anode having a much larger electron-collecting area that the physical anode. The voltage drop across the pre-anode double-layer can be as high as 30 Volts and is thus greater than the ionization potential of the process gas (see probe measurements in ¹⁶). Electrons accelerated in the double layer can ionize gas atoms or molecules inside the double layer or in the layer-enclosed ball-shaped volume which is attached to the anode surface. Experimentally, this volume is visible as a "plasma ball".

The next step is to go from stagnant gas to a gas flow. The gas is introduced through an opening in the cathode, and the small anode area is redesigned to form a nozzle letting the relatively dense plasma escape (Fig. 2(b)). The anode is "hollow" which lead Miljevic to name the discharge "hollow anode discharge" ¹⁷. This name misleadingly suggests that there is a hollow-anode effect in analogy to the well-known hollow-cathode effect. However, there is no hollow-anode effect; it is the smallness of the anode area which causes the appearance of a virtual anode and dense "plasma ball".

The geometry of the anode has a number of important implications. First, it represents a nozzle in which the plasma is gasdynamically accelerated up to supersonic velocities ¹⁶. The nozzle separates a region of relatively high pressure inside the source from relatively low pressure of the vacuum process chamber. The high pressure inside the source is necessary to

maintain the glow discharge (mean free path of electrons is smaller than characteristic discharge dimension), and the low pressure in the process chamber is required for MBE or similar deposition processes ¹⁸. A second implication of the geometry is that the pressure in the process chamber can be chosen in a wide range by a relatively small change in the nozzle diameter. A third implication is a low ion energy of the plasma flow. Ions born on the cathode-side of the double layer are blocked from escaping by the electric field of the double layer. According to the model, only ions formed on the anode-side of the double layer may escape, and their acceleration should be determined by gasdynamics. In the present work, ion energy distributions are measured to elucidate this point. The fourth implication is that material sputtered from the cathode surface has a vastly greater probability of being deposited inside the source chamber than escaping through the nozzle opening; the plasma is of high purity which allows its application to the growth of semiconductors. Finally, the fifth implication is that the expanding plasma plume contains not only ionized gas atoms or molecules but a large fraction of excited species and radicals which play an important role in the formation of compound films.

So far, the nozzle functioned as the anode, which lead to the name "hollow-anode plasma source". It has been found that the anode can also be located downstream of the nozzle; the nozzle part being at floating potential or made from insulating material such as quartz or ceramic. In this case, the previous name does not apply, and we have named the source "constricted-plasma source". The operation principle remains the same, namely a double layer forms at the high-pressure side of the constriction (nozzle) which facilitates current transport though the small cross section, and a flow of plasma is produced. In this sense, the hollow-anode plasma source is one version or mode of operation of the constricted-plasma source.

The source performance can be influences by a magnetic field. In some of the investigation, a CoSm permanent ring magnet was attached to the anode plate so that the nozzle and its downstream area are subject to an axial magnetic field. The highest field strength was 200 mT at the center of the ring axis.

III. PLASMA DIAGNOSTICS

The plasma stream exiting the source was investigated using Langmuir probes, optical emission spectroscopy, and a plasma monitor system (VG Quadrupoles SXP Elite). The latter is particularly useful because it allows mass-selective flux and energy measurements. The device entrance was a 1 mm diameter orifice in 10 cm distance from the nozzle. The variable parameter space includes kind of gas, gas flow rate, discharge current, geometrical factors such as pump location and pumping speed, location of measurements, etc. We report here only on selected measurements and limit the investigation to nitrogen as the discharge gas. We used a 0-1 kV power supply with a 3 k• external resistor.

Previous experiments have shown that in particular three parameters influence the plasma: the gas flow, the discharge current, and the distance from the source. Using the plasma monitor, the flux of atomic and molecular nitrogen ions was measured as a function of the discharge current and gas flow rate (Figs. 3 and 4). One can see that both ion fluxes behave similarly; the ratio of N^+/N_2^+ is roughly 1/4. The ion energy distribution, which is important for the growth of thin crystalline films, is shown for atomic nitrogen ions (Fig. 5) and molecular ions (Fig. 6). The distributions show two peaks. As anticipated, a large fraction of ions has a very low energy of a few eV. However, we do not have yet a clear understanding of the physical mechanisms causing the two-peak distribution. We have to look at the exact shapes of the distributions with some caution because we noticed that they

can be influenced by varying the ion optics inside the plasma monitor. Nevertheless, it is safe to say that most ions have a kinetic energy of 20 eV or less. This result is important when compared to the much higher ion energies of other plasma and ion sources (usually 30 eV and higher).

The situation is somewhat different when the constriction is floating or insulating and the anode is remotely located from the source. In this case, most the total anode-cathode voltage is between in the constriction and anode, and ions of higher energy (up to 100 eV) have been found. In this case the constriction electrode causes not only a double layer on the high-pressure side but acts like a hollow cathode for the discharge plasma on the low-pressure side. Ion are accelerated in the "cathode fall" which forms on the low pressure side. This interpretation is supported by the fact that a Negative Dark Space appears at low flow rates (5 sccm or less).

Optical emission spectra show the characteristic bands of the N_2 and N_2^+ . The spectra were measured side-on, i.e., with the optical axis perpendicular to the source and flow axis. Specifically, we measured the intensity of the First Positive System (FPS), $B^3\Pi_g \to A^3\Sigma_u^+$, and the Second Positive System (SPS), $C^3\Pi_u \to B^3\Pi_g$, of the (neutral) nitrogen molecule, and the First Negative System (FNS), $B^2\Sigma_u^+ \to X^2\Sigma_g^+$, of the ionized molecule. The energetics of the plasma can be displayed, for instance, by the intensity ratio of the less-energetic first positive system and second (more energetic) positive system whose upper electronic levels are 7.3 eV ($B^2\Sigma_u^+$) and 11.1 eV ($C^3\Pi_u$) above the molecule's ground state ¹⁹. We found that this ratio increases with increasing flow rate. Figure 7 shows the dependence of the SPS (neutral molecule) and FNS (molecular ion); also here we see that less ions are produced or excited when the flow increases beyond a certain rate. These

observations are consistent with probe and plasma monitor measurements which show that the ionization efficiency becomes smaller at high gas flow rates. Langmuir probe measurements of electron density and temperature have been done at various distances from the source. For example, at 5 cm distance and with a current of 100 mA, $n_e = 7 \times 10^7$ cm⁻³ at a flow of 5 sccm, dropping to $n_e = 4 \times 10^7$ cm⁻³ at 100 sccm. For the same example, the electron temperature drops from $T_e = 8$ eV (5 sccm) to $T_e = 6$ eV (100 sccm).

IV. SUMMARY

- 1) The Constricted Plasma Source produces a flux of low energy ions (<20 eV) in the hollow-anode mode but contains higher-energy ions when operated with a floating or insulating nozzle. The low ion energy is consistent with the successful growth of high-quality crystalline GaN using the source in the hollow-anode mode ²⁰.
- 2) The plasma flow contains a mixture of various species, its composition depends mainly on the gas flow, discharge current, and distance from the source. The degree of ionization and excitation decreases with increasing gas flow and increases with increasing discharge current.
- 3) The ratio of atomic nitrogen ions to molecular nitrogen ions is roughly of order 1/4 and depends slightly on various parameters. The flow contains a significant but not well known fraction of dissociated and excited molecules which contribute to reactive film growth. The density of activated nitrogen (ionized, dissociated and excited species) drops considerably with distance from the source.

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REFERENCES

- H. R. Kaufman and R. S. Robinson, in *Handbook of Ion Beam Processing Technology*, edited by J. J. Cuomo, S. M. Rossnagel and H. R. Kaufman (Noyes, Park Ridge, 1989), pp. 8-20.
- W. C. Hughes, W. H. Rowland, M. A. Johnson, S. Fujita, J. W. Cook, J. F. Schetzina, J. Ren, and J. A. Edmond, J. Vac. Sci. Technol. B 13, 1571-1577 (1995).
- A. Farachi, J. Aubert, L. Wartski, P. Coste, and C. Mardirossian, Nucl. Instrum. Meth. Phys. Res. B **74**, 457-459 (1993).
- W. M. Holber, in *Handbook of Ion Beam Processing Technology*, edited by J. J. Cuomo,
 S. M. Rossnagel and H. R. Kaufman (Noyes, Park Ridge, 1989), pp. 21-38.
- R. S. Robinson and H. R. Kaufman, in *Handbook of Ion Beam Processing Technology*, edited by J. J. Cuomo, S. M. Rossnagel and H. R. Kaufman (Noyes, Park Ridge, 1989), pp. 39-57.
- ⁶ Y. Rao, J. Wu, and T. Xu, Rev. Sci. Instrum. **67**, 3494 (1996).
- A. Anders, R. A. MacGill, I. G. Brown, and A. Vizir, 7th Int. Conf. Ion Sources, Taormina, Italy, 1997

- M. v. Ardenne, *Tables of Electron Physics, Ion Physics and Electron Microscopy* (Deutscher Verlag der Wissenschaften, Berlin, 1956).
- 9 R. Weiss, C. Winkler, and R. W. Schrittwieser, Plasma Sources Sci. Technol. **6**, 247-249 (1997).
- R. W. McCullough, in *Application of Accelerators in Research and Industry*, edited by J.
 L. Duggan and I. L. Morgan (AIP Press, New York, 1997), pp. 275-278.
- 11 N. Xu, Y.-C. Du, Z.-F. Ying, and F.-M. Li, Appl. Phys. Lett. **69**, 1364-1366 (1996).
- 12 E. C. Samano, W. E. Carr, M. Seidl, and B. S. Lee, Rev. Sci. Instrum. 64, 2746-2752 (1993).
- 13 V. I. Miljevic, Appl. Optics **23**, 1598-1600 (1984).
- 14 V. I. Miljevic, Rev. Sci. Instrum. **61**, 312-314 (1990).
- 15 V. I. Miljevic, Radiat. Phys. Chem. **35**, 667-669 (1990).
- 16 A. Anders and S. Anders, Plasma Sources Sci. Technol. 4, 571-575 (1995).
- 17 V. Miljevic, Rev. Sci. Instrum. **55**, 931-933 (1984).
- A. Anders, N. Newman, M. Rubin, M. Dickinson, A. Thomson, E. Jones, P. Phatak, and A. Gassmann, Rev. Sci. Instrum. 67, 905-907 (1995).
- 19 A. Lofthus and P. H. Krupenie, J. Phys. Chem. Ref. Data 6, 113-307 (1977).
- 20 M. S. H. Leung, R. Klockenbrink, C. Kisielowski, H. Fujii, *et al.*., Mat. Res. Soc. Symp. Proc. **449**, 221-226 (1997).

FIGURE CAPTIONS

Fig. 1 Construction of the constricted-plasma source (source length 6.3 cm without optional magnet, 8.6 cm with magnet; diameter 6.6 cm; constriction diameter 1 mm).

- Fig. 2 Formation of a virtual anode. (a) Case of a small anode area. (b) Case of a constriction with gas flow.
- Fig. 3 Flux of atomic nitrogen ions (mass number 14) as a function of the discharge current with the gas flow as a parameter. The units are counts of the plasma monitor detector.
- Fig. 4 As Fig. 3 but for molecular ions (mass number 28).
- Fig. 5 Energy distribution function of atomic nitrogen ions at a flow rate of 10 sccm, with the discharge current as a parameter. Only every 10th data point is indicated for better readability.
- Fig. 6 As Fig. 5 but for molecular nitrogen ions.
- Fig. 7 Intensity of the Second Positive System (various vibrational transitions) and the First
 Negative System (vibrational transition 0 → 0) as a function of the gas flow rate.
 Data are normalized to their value at 100 sccm; discharge current 100 mA;
 measurements at 8.5 cm distance form source.